Mechanisms of lightning formation in deep maritime clouds and hurricanes (The HAMP contribution)

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Abstract

In maritime clouds over warm surface most droplets formed at the cloud base fall out without ever reaching the freezing level. Nevertheless, lightning takes place sometimes in the eye walls of hurricanes, where clouds are, supposedly, the most maritime in the world. In this study we address the following question: "Why does lightning take place in deep maritime convective clouds in the Intertropical Convergence zone and in hurricane eyewalls at all?"

Numerical simulations using the spectral microphysics Hebrew University cloud model show that the formation of lightning in maritime clouds requires two conditions to be satisfied: a) significant vertical velocities and a large cloud-depth, and b) the existence of small aerosols with the radii lower than about $0.05\ \mu m$ in diameter in the cloud condensational nuclei (CCN) size spectra. Both factors are necessary components for the lightning to occur. Any absence of small aerosols would prevent in-cloud nucleation of small drops at high distances above the cloud base. Any lack of sufficiently fast updrafts prevents in-cloud nucleation and the formation of supercooled water at high levels. Small aerosols form over the ocean, supposedly, due to chemical reactions followed by particle collisions to create the aerosol accumulation mode. Small aerosols can be of continental origin and may penetrate into oceanic regions, like sulphate

pollution or African dust. The latter may explain the existence of intense lightning in the ITCZ over the ocean near the African coast. The potential importance of small CCN in creation of supercooled water at the upper levels, high ice crystals concentrations in cloud anvils and in other microphysical features of deep tropical convection is discussed.

1. Introduction

Observations indicate that the concentration of maritime cloud condensational nuclei (CCN) (at 1% supersaturation) is about 60-200 cm^{-3} (Pruppacher and Keltt 1997; Levin and Cotton 2009). It is widely accepted that aerosol size distributions over the sea contain higher concentrations of giant CCN (hereafter GCCN) with dry radii exceeding about 1 μm than those over the land because of their production by sea spray. It is usually assumed that low CCN concentrations and the existence of giant CCN determines typical maritime properties of these clouds (especially tropical clouds), characterized by very low droplet concentrations of about 50 cm^{-3}, and rapid formation of raindrops below the freezing level. The raindrops, in turn, collect most of the drops nucleated near the cloud base, leading to intense warm rain at the surface. Raindrops that did not fall out below 4-5 km level freeze within the layer between 5 and 6 km altitude, forming a comparatively low concentration of graupel. Such a view of maritime clouds is widely accepted in the literature (see reviews by Rosenfeld et al, 2008; Khain et al 2009). According to this view maritime tropical cloud should not contain any significant supercooled water content at high (~8-10 km) levels. For instance, one of the reasons for the termination of the STORMFURY (1965-1975) project (according to which intensification of deep convection of the eyewall periphery of tropical cyclones supposed to be reached by glaciogenic seeding with AgI of cloud anvils) was the low amount of supercooled water and high concentrations of natural ice crystals at the upper levels in these clouds as it was supposed by Willoughby et al (1985).

On the other hand, some observations in deep maritime convection indicate that the microphysical structure of marine deep convection is more complicated than that described above.

The first piece of evidence is the existence of bimodal droplet size distributions in maritime clouds at a distance of a few km above cloud base (e.g. Warner 1969a,b). The radii of droplets belonging to the second cloud mode that arises 1-2 km above cloud base ranges typically from 3 μm to about 10 μm , while the droplets forming the first mode have radii ranging from 10 μm to 20 μm. [Note that the droplet size distributions (hereafter, DSD) in deep continental convective clouds are more frequently unimodal (Pinsky and Khain 2002) . The importance of this fact will be discussed below].

Another phenomenon which indicates the existence of supercooled water at cold temperatures in deep maritime clouds is the existence of lightning in tropical cyclone (TC) eyewalls and maritime deep convection in the Intertropical Convergence Zone (ITCZ). According to a widely accepted concept the charge separation in clouds takes place in the zones of low temperatures (about -15 ^oC to -20 ^oC), where collisions between low and high density ice take place in the presence of a significant amount of supercooled water (e.g., Black and Hallet 1999; Takahashi, 1978; Saunders 1993, Cecil et al 2002a,b; Sherwood et al 2006; Reynolds *et al.* 1957; Takahashi 1978, 1984; Jayaratne *et al*. 1983; Baker *et al*. 1987; Helsdon and Farley 1987; Latham and Dye 1989; Kumar and Saunders 1989; Norville *et al*., 1991; Saunders *et al.* 1991; Helsdon *et al.* 2001, 2002). The significant difference in the lightning density over the land and the sea is well known (e.g., Orville and Henderson 1986; Christian and

Latham 1998; Williams and Satori, 2004; Williams et al 2004, Sherwood et al 2006). The lower frequency of lightning over the sea is usually attributed to low vertical velocities in maritime convection (Black et al 1996; Szoke et al 1986; Jorgensen et al 1985; Williams et al 2004, 2005) and low aerosol concentrations (Takahashi 1984; Molinie and Pontikis 1995; Rosenfeld and Lensky 1998; Michalon *et al.* 1999; Williams *et al.* 1999, 2002; McCollum *et al.* 2000; Williams and Stanfill 2002). Both factors favor the formation of raindrops below freezing level collecting most of the small droplets nucleated near the cloud base. This effect should dramatically decrease the amount of supercooled water droplets aloft, tending to inhibit the charge separation process. At the same time **Figures 1 and 2** show satellite observations of intense lightning in deep maritime clouds over the open sea including extremely maritime clouds within the TC eyewall. If the concept of lightning formation is correct, such observations imply that at upper levels supercooled droplets should exist in extremely maritime clouds.

At last, amounts of supercooled water of ~0.3 gm^{-3} were observed by aircraft in deep convective updrafts with peak vertical velocities of up to 20 m/sec in The Cirrus Regional Study of Tropical Anvils and Cirrus Layers - Florida Area Cirrus Experiment (CRYSTAL-FACE) over the sea near Florida (Fridlind et al. 2004; Heymsfield et al. 2005; Phillips et al. 2005). Heymsfield et al. estimated that updraft speeds faster than a few metres/sec were needed to support a positive supersaturation with respect to water and survival of supercooled liquid during ascent throughout the mixed-phase region (0 to -36 ^{o}C), in parcel model simulations of CRYSTAL-FACE updrafts. Moreover, aircraft observations in the Kwajalein Experiment (KWAJEX) showed droplet concentrations of about 50 cm^{-3} up to about 9 km altitude (about -30 ^{o}C) throughout most of the depth of the mixed-phase region, averaged over deep, highly visible cloud-decks. These microphysical features were simulated by Phillips et al. (2007a) with a cloud-system resolving model with double-moment bulk microphysics and an interactive CCN and IN components.

Note another phenomenon that hardly can be explained without assuming the existence of a significant amount of small cloud droplets in convective updrafts supplying anvils of deep maritime cumulonimbus clouds. It is known that the optical depth of anvils of deep convective clouds in the ITCZ is quite significant. This is consistent with the existence of concentrations of about 1 cm^{-3} of small ice crystals in the cloud anvils. Such concentrations observed in situ by Takahashi and Kuhara (1993) exceed by several orders of magnitude the possible concentration of active ice nuclei (IN) in those remote maritime regions, and can be attributed to homogeneous freezing of droplets at temperatures as cold as -38 ^{o}C (Rosenfeld and Woodley, 2000; Khain et al 2001; Phillips et al. 2005, 2007a).

Hence, in this study we address a question, which is just opposite to that usually asked, namely: "If warm rain processes in maritime clouds are so efficient, then why can lightning form in deep maritime convective clouds in the ITCZ (Fig 1, red circles) and in the hurricane eyewalls (Fig 2) at all?" Why do supercooled droplets exist so high aloft in maritime clouds with moderate updrafts?

In the present study, we argue that all these phenomena are caused by one and the same mechanism of in-cloud droplet nucleation, when the supersaturation in ascending cloud parcels exceeds the local maximum at the cloud base. Such possibility was discussed in some previous studies (Ochs 1978; Ludlam 1980; Song and Marwitz 1989; Korolev 1994; Pinsky and Khain 2002; Segal et al, 2003; Phillips et al. 2005). Such a rise in supersaturation can be caused by an increase in the vertical velocity above the cloud base and the depletion of cloud-droplets by coalescence during deep maritime ascent (Ochs 1978; Pinsky and Khain 2002; Phillips et al

2005). Pinsky and Khain (2002) showed that the combined effects of vertical velocity increase and the depletion of cloud droplets by coalescence can lead to formation of DSDs containing three modes.

Note that the hypothesis of in-cloud nucleation in air ascending from the cloud base requires the existence of very small aerosol particles (small CCN) with diameters smaller than about 0.05 μm, which can be activated only at high supersaturations significantly exceeding 1%. These AP hardly can be activated at the cloud base of maritime clouds because of a relatively low supersaturation there. Besides, these AP can scarcely be scavenged by precipitation because of their small sizes and very low scavenging rate (Flossmann and Pruppacher 1987). As a result, both droplets and haze particles (not activated CCN) ascend within updrafts above cloud base. Decreasing drop concentrations by collisions and an increase in the vertical velocity above cloud base may together cause activation of these haze particles, as noted above, forming droplets several km above the cloud base as the supersaturation is boosted (Ochs 1978; Pinsky and Khain, 2002; Phillips et al 2005).

So, the explanation of the phenomena listed above reduces to answering the following questions:

a) "Do small APs with diameters below 0.03-0.05 μm exist in the maritime tropical atmosphere, and if yes, what is their concentration?"

b) Is the supersaturation in deep maritime clouds high enough to activate these smallest APs?

[Note that there is another possible mechanism of in-cloud nucleation at high levels, namely, lateral entrainment of free-tropospheric air containing inactivated CCN (Fridlind et al, 2004; Phillips et al 2005, 2007a). This mechanism requires is not considered in the present study]

According to Twomey and Wojciehowsky (1969), Hegg and Hobbs (1992), Hegg et al (1993); Pruppacher and Keltt (1997) and Levin and Cotton (2009) the concentration of activated CCN in the maritime atmosphere increases monotonically with the increase in supersaturation up to the values as high as 8-10% (**Figure 3**). The critical diameter of soluble AP that can be activated at supersaturation of 10 % is about 0.012 μm. It is a general practice to describe the dependence of cloud nucleation nuclei (CCN) concentration using a semi-empirical formula

$$N_{ccn} = N_o S^k , \qquad (1)$$

where N_{ccn} is the concentration of activated AP at supersaturation S (in %) with respect to water, N_o and k are the measured parameters. Parameter k is known as the slope parameter. The values of k reported over the ocean vary from 0.3 to 1.3 within a wide range of supersaturations in different zones of the ocean (see Pruppacher and Keltt 1997), and even within different air masses in the same geographical location (Hudson and Li 1995). The large values of k at high supersaturations indicate the existence of a significant amount of small aerosols in the maritime atmosphere.

At the same time, in some observational (e.g., Hudson 1984; Hudson and Frisbie 1991; Hudson and Li 1995; Hudson and Yum, 1997; 2002) and laboratory studies (Jiusto and Lala 1981) a decrease in the value of k with increasing of supersaturation was reported. According to these results no new CCN can be activated at supersaturations exceeding some threshold S_{thr} which is assumed to vary from 0.1 % (Cohard et al, 1998; Emde and Wacker 1993) to about 0.6% (Hudson and Li 1995). Such observations connote a lack of small APs with dray diameters of about 0.05-0.1 μm or less in the sampled air, as seen sometimes for example in the remote maritime boundary layer (e.g., Clarke and Kapustin 2002). The $k(S)$ dependence with the condition that $k \approx 0$ at $S > S_{thr}$ is used in several studies (e.g., Cohard et al 1998; Abdul-Razzak et al. 1998) for parameterization of aerosol activation in cloud models. **Figure 3** depicts the variability of $N_{ccn}(S)$ dependencies reported for maritime aerosols in different studies. The existence (absence) of S_{thr} of about 0.6% indicates the lack (presence) of CCN aerosols with diameters below ~0.1 μm in the maritime atmosphere.

The discussion during a meeting dedicated to the WMO/IUGG scientific review processing (France, Toulouse, October 2006) showed that the question about the existence of small condensational nuclei in the maritime atmosphere remains open. Despite this evident uncertainty, the observations of bimodal DSD spectra, lightning in the TC eyewall, and high ice crystal concentrations and optical depth of deep tropical cloud anvils are consistent with the existence of small APs that can be activated under high supersaturations. Indeed, such small aerosols have been directly observed in the free troposphere over the remote Pacific (Clarke and Kapustin 1992). The existence of small AP follows also from the AP budget. According to results of many studies (Twomey, 1968, 1971; Radke and Hobbs, 1969; Dinger et al., 1970; Hobbs, 1971; Levin and Cotton 2009) sea spray contributes mainly the large AP tail of the AP size distribution, but most CCN in the accumulation mode are formed by collisions of smaller aerosols having another source different from the sea spray. The small AP can be of continental nature (like Saharan dust, which is often was found in convective storms near the Eastern African coast and in storms and hurricanes reaching the American coast) or can form via different chemical reactions over the sea (Pruppacher and Klett 1997, Clark and Kapustin 2002).

Many numerical simulations of aerosol effects on cloud microphysics, dynamics and precipitation (e.g., Khain 2004, 2005, 2007; Khain 2009; van der Heever et al 2006, Wang et al 2005, Tao et al 2007, Phillips et al. 2001 ,2002, 2005, 2007, 2009) have been carried out under different AP concentrations typical of maritime and continental conditions. In most studies parameter N_o is usually varied within a wide range from a few tens to several thousand of AP per cm^3. The role of the slope parameter is typically not discussed, and implicitly its role is assumed not to be decisive. In some of these studies the slope parameter was assumed to decrease with increasing supersaturation, which substantially decreased the efficiency of in-cloud nucleation. Besides, in many bulk-parameterization microphysical schemes and cloud parameterization used in global circulation models nucleation is assumed to be restricted to the cloud base.

As will be shown in the present study, small aerosols can dramatically modify maritime convection, where the supersaturation in clouds can be very high in fast or precipitating convective updrafts. Small aerosol particles can be activated into droplets.

2. Model description

The HUCM is a 2-D mixed-phase model (Khain and Sednev 1996; Khain et al 2004, 2005, 2008a,b) with spectral bin microphysics based on solving a system of kinetic equations for size distribution functions for water drops, ice crystals (plate-, columnar- and branch types), aggregates, graupel and hail/frozen drops, as well as atmospheric aerosol particles (AP). Each size distribution is described using 43 doubling mass bins, allowing simulation of graupel and hail with the sizes up to 5 cm in diameter. The model is specially designed to take into account the AP effects on the cloud microphysics, dynamics, and precipitation. *The initial* (at t=0) CCN size distribution is calculated using the semi-empirical dependence (1) and applying the procedure described by Khain et al (2000). At t>0 the prognostic equation for the size distribution of non-activated AP is solved. Using the supersaturation values, the critical AP radius is calculated according to the Kohler theory. The APs with the radii exceeding the critical value are activated and new droplets are nucleated. The corresponding bins of the CCN size distributions become empty.

Primary nucleation of each type of ice crystals is performed within its own temperature range following Takahashi et al (1991). The dependence of the ice nuclei concentration on supersaturation with respect to ice is described using an empirical expression suggested by Meyers et al. (1992) and applied using a semi-lagrangian approach (Khain et al 2000) allowing the utilization of the diagnostic relationship in the time dependent framework. The secondary ice generation is described according to Hallett and Mossop (1974). The rate of drop freezing is described following the observations of immersion nuclei by Vali (1975, 1994), and homogeneous freezing according to Pruppacher (1995). The homogeneous freezing takes place at temperature about -38^{o}C. The diffusion growth/evaporation of droplets and the deposition/sublimation of ice particles are calculated using analytical solutions for supersaturation with respect to water and ice. An efficient and accurate method of solving the stochastic kinetic equation for collisions (Bott, 1998) was extended to a system of stochastic kinetic equations calculating water-ice and ice-ice collisions. The model uses height dependent drop-drop and drop-graupel collision kernels following Khain et al, (2001) and Pinsky et al (2001). Ice-ice collection rates are assumed to be temperature dependent (Pruppacher and Klett, 1997). Detailed melting procedure with calculation of liquid water fractions within melting aggregates, graupel and hail is included following Phillips et al (2007b). Advection of scalar values is performed using the positively defined conservative scheme proposed by Bott (1989).

3 The experimental design

All runs were performed with a 2D computational domain is 178 km x 16 km, and a resolution of 250 m and 125 m in the horizontal and vertical directions, respectively.

To show the potential effect of smallest aerosols on cloud microphysics and precipitation, a set of deep convective clouds were simulated under thermodynamical conditions typical of tropical oceans during hurricane season (Jordan 1958). The sounding indicates 90 % humidity near the surface. The freezing level is at 4.2 km. The atmosphere is relatively unstable under these conditions, so the maximum vertical velocities in the simulated clouds ranged from 15 to 18 m/s. It means that these convective clouds fall into the range of the 5% of the most intense maritime clouds according to Jorgensen et al (1985) and Jorgensen and LeMone (1989). The initial CCN size distribution was calculated using Eq. (1) where N_o was assumed equal 100 cm^{-3} in all simulations. The method of calculation of initial DSD using Eq. (1) is described by Khain et al (2000). Sensitivity simulations were performed with respect to the slope parameter k which was assumed equal to 0.3; 0.6 and 0.9. These simulations were identical in all other respects. The specific feature of the simulations as compared to those reported in Khain et al (2005, 2009) was that the minimum AP diameter was assumed equal to 0.006 μm, which corresponds to the minimum size in the accumulation mode. We suppose that there no AP smaller than this size. AP with diameter of 0.006 μm can be activated at supersaturations of ~40%. Based on the data from Pruppached and Klett (1997) concerning the range of typical values of slope parameter over the ocean, these simulations will be referred to as the "weak-slope" case, “mean-slope” and "steep-slope" cases for k = 0.3, 0.6 and 0.9 respectively.

Corresponding dependencies of concentration of CCN on supersaturation are shown in **Figure 3**.

The maximum dry AP diameter in the model was assumed to be 4 μm. The largest of these particles are transformed to droplets with diameter of 16 μm at the cloud base. No GCCN with dry diameter larger than 4 μm were allowed.

3. Results of simulations

Figure 4 shows the fields of the droplet concentration N_d, the cloud (CWC) and rain (RWC) water contents respectively, in the simulations with different slope parameters. In all three simulations one can see several typical features common to all three simulations (which are typical of maritime clouds): the droplet concentration does not exceed 100 cm^{-3}; warm rain forms fast, mainly below the freezing level. Raindrops reach levels of ~ 5 km altitude above mean sea level (MSL).

However, there is a dramatic difference in the droplet concentration and the cloud water content above 5 km MSL. At *k*=0.3 the concentration rapidly decreases with height and the CWC above the freezing level is very small (less than 0.2 gm^{-3}). In the steep-slope case (*k*=0.9) the droplet concentration increases with altitude above z=6 km indicating intense in-cloud nucleation aloft. Large supercooled LWC with maximum of 1.8 gm^{-3} forms within the layer 6-9 km. The mean-slope case (*k*=0.6) indicates intermediate results. In-cloud nucleation and formation of supercooled water content with maximum of 0.8 gm^{-3} at z=8 km is seen. The supercooled water content rapidly decreases above the level due to freezing, including that caused by riming. The maximum vertical velocities in these simulations are 15-21 m/s. It is remarkable that maximum vertical updrafts take place in the steep-slope case (k=0.9) because of the extra latent heat release caused by diffusion growth of newly nucleated droplets.

Figure 5 shows droplet mass distributions at several height levels along cloud axes in the simulations for the weak- and speep-slope cases (k=0.3 and *k=0.9)* at t=1500 s. One can see that the DSDs are actually similar below 2.5 km MSL. However, a dramatic difference takes place above 5 km MSL. While in the weak-slope case (*k=0.3*) the LWC is negligible at z= 8.5 km (T=-20^{o}C), in the steep-slope (*k=0.9*) case the LWC is significant, and drop sizes range from 40 to 500 μm at this level. Note that the drop mass spectrum in case k=0.9 at z=7.5 km contains smaller drops (with the radii of 10 μm) than the spectrum at z=5 km. This indicates the nucleation of new droplets (in-cloud nucleation) within the layer from 5 to 7 km. In agreement with analysis by Pinsky and Khain (2002), an increase in the vertical velocity with height seen in **Figure 6** (left) leads to the increase in supersaturation to ~12 %, which substantially exceeds the supersaturation value near the cloud base (~0.5 %). (Note that comparatively crude model resolution of 125 m in the vertical direction does not allow one to resolve the peak of supersaturation that is typically only 20 m above cloud base in real clouds).

This increase in supersaturation is caused also by the decrease in the droplet concentration by efficient collisions. Note that the existence of such high supersaturations follows from the theory for an adiabatic parcel (e.g., Rogers and Yau 1989; Korolev and Mazin, 2003) according to which the equilibrium supersaturation which is rapidly reached in the presence of liquid droplets is proportional to the ratio of the vertical velocity to the droplet concentration. The combination of low drop concentration and high vertical velocity leads to activation of small aerosols (Pinsky and Khain 2002). According to the Kohler law at supersaturation of 12 %-15% soluble AP with dry diameters as low as 0.01 μm are activated. In simulation with k=0.3, the minimum size in the droplet spectra monotonically increases with height (**Figure 5** right). The minimum drop radius at z=7.5 km in this run is 100 μm. The latter does not show any pronounced in-cloud nucleation in this run because of lack of smallest aerosols.

Figure 7 shows fields of graupel and ice crystal contents in the weak- and steep-slope simulations (k=0.3 and 0.9) at t= 1800 s. One can see that in the steep-slope *case (k=0.9)* there is a large region within the cloud at temperatures below -13 ^{o}C, where graupel, ice crystals and supercooled droplets co-exist, which is considered as a favorable condition for charge separation and lightning formation. These conditions were found favorable for lightning formation. These conditions were found favorable for lightning formation in the TC eyewall (e.g., Black et al 1996). In the weak-slope case (k=0.3) conditions for lightning are unfavorable in spite of high vertical velocities and supersaturations because of the negligible amount of supercooled water.

Note that mass of crystals in the weak-slope simulation (k=0.3) turned out to be larger than in steep-slope case (k=0.9). It can be attributed to the formation of supercooled water reducing the supersaturation in the steep-slope case, so that supersaturation where crystals exist is higher in the weak-slope case (k=0.3). At the same time concentration of crystals above the level of homogeneous freezing (~10 km) is significantly (by factor of 60) higher in the steep-slope case (k=0.9) and reaches in maximum $50\,cm^{-3}$. These crystals are formed by homogeneous freezing of small droplets formed by in-cloud nucleation near cloud top (Figure 4, upper row). In the mean-slope simulation (k=0.6) the concentration of ice crystals forming in cloud anvil by homogeneous droplet freezing is about $20\,cm^{-3}$. In case of no small aerosols allowed (when no nucleation is assumes to occur at supersatiration larger than 0.6 %) concentration of ice crystals in the cloud anvil is determined by primary nucleation and is about 40-50 l^{-1}, and is not related to homogeneous droplet freezing. Thus, according to the results of our simulations, the existence of small AP is of crucial importance for formation of high ice crystal concentrations needed to explain high optical depth of cloud anvils seen in the ITCZ.

To investigate the role of the vertical velocity (atmospheric instability) a supplemental simulation has been performed which differed from that with k=0.9 by more stable sounding under which the maximum vertical velocity was about 7 m/s. This speed is 2-3 times less than that in the standard k=0.9 run. **I**n this case, cloud top height is lower than in the standard steep-slope case. The supercooled LWC is reduced as well and concentrates largely within the layer between the freezing level and the 5.5 km height level. Concentrations of ice crystals and graupel within this layer are quite low. It means that in regions of weak vertical velocity typical of many maritime clouds lightning, as well as high ice crystal concentrations can hardly form at all. This is true even when there are copious small aerosols with diameter below 0.05 μm.

4. Conclusion and discussions

This study demonstrates the importance of atmospheric aerosols with diameters below ~0.03-0.05 μm in the creation of microphysical structure of deep convective clouds over the ocean. Low CCN concentration (at supersaturation of 1%) determines rapid formation of warm rain and decrease in concentration of droplets nucleated at cloud base. Under such conditions supersaturation in cloud updrafts of maritime convective clouds increases during ascent. Another factor contributing to the rise in supersaturation is the vertical acceleration of the updraft during ascent due to its positive buoyancy. These factors may lead to in-cloud nucleation and formation of small cloud droplets several km above the cloud base. The existence of giant CCN and other factors accelerating warm rain formation cannot prevent formation of new droplets aloft. Moreover, the faster is washout due to warm rain, the higher can be supersaturation in clouds aloft and the greater the production of supercooled droplets at upper levels. The process of in-cloud nucleation allows one to explain formation of bimodal droplet spectra in maritime clouds, formation of lighting in extremely maritime clouds in eyewall of hurricanes, high optical depth of anvils of deep tropical clouds in the ITCZ.

To be efficient, all these mechanisms require two conditions: the existence of a significant amount of AP with diameters below ~0.03-0.05 μm, and a significant vertical velocity. In maritime clouds in environments with CCN concentrations below 100 cm^{-3} (at 1% supersaturation) an appreciable vertical velocity is required for creation of high in-cloud supersaturations that exceed that at cloud base (according to the calculations, this supersaturation at levels of 6 km may exceed 10-12%) that is able to activate smallest aerosols well above cloud-base. The lack of high updrafts will prevent in-cloud nucleation and the formation of supercooled water at high levels, where co-existence with graupel and crystals would be possible.

Khain et al (2008b) investigated effects of continental aerosols ingested into deep maritime clouds of TC during TC landfall. It was shown that the direct penetration of a large amount (~1500 cm^{-3}) continental aerosols with sizes exceeding ~0.01 μm into the TC also leads to formation of a significant amount of supercooled water content. These AP nucleate to droplets at cloud base. In case the concentration of the AP is high, clouds get "continental" properties, the production of warm rain is less intense and a significant number of supercooled droplets reaches the upper levels. We see, therefore, that the same effects, namely, production of supercooled water at the upper part of clouds and significant amount of ice crystals in cloud anvils can be reached via two different mechanisms: “maritime” one, based on in-cloud nucleation, and a “continental” one based on aerosol-induced reduction of the warm rain and penetration of small droplets from cloud base to the upper levels.

Note that both mechanisms require significant vertical velocity in clouds to be efficient. It seems, that the maritime and continental mechanisms tend to oppose each other. The “continental” mechanism, by keeping the droplet number mixing ratio relatively high till the upper levels, tends to decrease supersaturation aloft in clouds, which decreases the extent of in-cloud nucleation (or just prevents it). To be efficient, the continental mechanism does not need the smallest AP. The comparative contribution of these two aerosol effects on cloud microphysics depends on the concentration of “continental aerosols”. When the concentration of continental CCN exceeds ~1500-2000 cm^{-3}, the role of the continental mechanism seems to prevail (it does not exclude the possibility of in-cloud nucleation aloft, but the concentration of newly nucleated droplets will be lower than that of droplets penetrating from the cloud base).

It may be hypothesized that lightning in eyewall and at the periphery of a landfalling TC seen in Figure 2 is caused by different mechanisms: in the eyewall the “maritime” mechanism is efficient, while at the TC periphery “continental” mechanism (suppression of warm rain) may dominate.

It seems that the role of giant CCN turns out to be not crucial in both cases. In “maritime” mechanism GCCN cannot prevent (or can rather favor) the formation of high supersaturation fostering in-cloud nucleation aloft. The intense lightning in eyewalls of hurricanes is often observed at the mature TC stage, when the production of sea spray and GCCN is especially intense. In “continental” mechanism high droplet concentration decreases the supersaturation in cloud updrafts which makes the growth of drops forming on GCCN slow. Smaller droplets growing faster may produce raindrops by autoconversion in amounts substantially exceeding the amount of GCCN (if the GCCN concentration is relatively low). The role of GCCN should be investigated in numerical simulations in more detail.

The important role of smallest aerosols in microphysics of maritime clouds found in the present study imposes heavy demands on the measurements of aerosols in the maritime atmosphere. The range of supersaturation variation in the laboratory should be increased to measure concentration of particles that can be activated at supersaturations as high as 10 % and even higher. The vertical variation of their concentration throughout the depth of the troposphere must also be measured.

One can assume two main sources of the small CCN over the ocean. One source is related to chemical reactions followed by collisions between APs to create the accumulated mode. Clarke and Kapustin (2002) analyzed aerosol observations from field campaigns over the Pacific Ocean and reported the existence of ultrafine aerosols with sizes smaller than 0.01 μm in the upper troposphere, especially near the outflow from deep convection (e.g. near the ITCZ). They assumed that these small APs, apparently generated in the outflow aloft, would be expected to subside through the free troposphere and may be eventually entrained into the boundary layer and into deep clouds through their lateral boundaries. The important role of such entrainment was stressed by Fridlind et al. (2004) and Phillips et al. (2005).

We also speculate that small aerosols can be of continental nature and penetrate the ocean with the intrusion of African dust. For instance, Hudson and Yum (2002) show (Figure 1) that the concentration of small aerosols is very low under clean Arctic conditions, while it is high enough in the Florida maritime air masses. The latter can explain the existence of intense lightning in the ITCZ near the African coast (Chronis et al 2007). The analysis of the lightning map (Figure 1) indicates a significant lightning downwind of continents, including in regions where the ocean surface is cooler due to ocean currents (e.g. off the western coast of the USA), which must tend to lower the atmospheric instability. To answer the question as regards the existence of small aerosols in the maritime tropical atmosphere more microphysical measurements of deep marine clouds and aerosol spectra in the zones of lightning over the ocean are required. The existence of the bimodal cloud droplet spectra in zones of high updrafts would be consistent with the existence of small aerosols.

Note that the role of small aerosols in the maritime atmosphere is not limited to their effects on lightning. The lightning serves in this case as just an indicator of the existence of a certain microphysical cloud structure. If the role of small aerosols is as important as suggested by the study, many concepts concerning the role of sea spray, giant CCN, etc. can be reconsidered, at least as concerns the microphysics of deep convective clouds developing over the oceans.

Note in conclusion that the effect of in-cloud nucleation discussed in the study could not be found in many numerical models with bulk microphysics because they perform cloud nucleation at the cloud base only. We suppose that description of in-cloud nucleation has to be included in the meteorological models (for instance, as done in the double-moment bulk scheme by Phillips et al.2007a, 2009) for better understanding aerosol effects. Moreover, the conclusions reached as regards the aerosol effects on precipitation should be possibly revised taken into account the role of smallest aerosols.

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List of figures

Figures

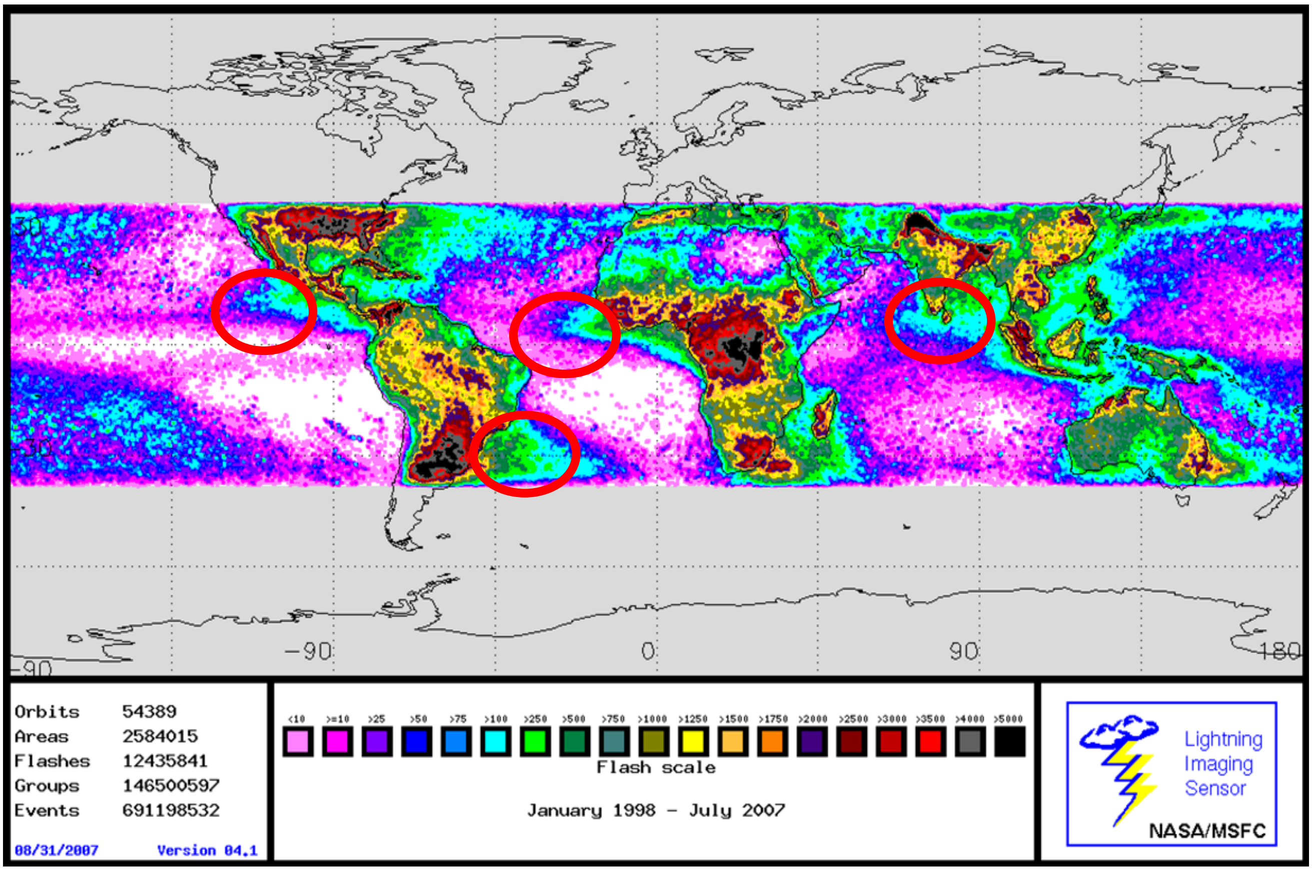

Figure 1. Global Lightning Distribution for the period from January 1998 to July 2007. One can see that intense lightning over the ocean (marked by ellipses) is observed in regions located downwind from the continents (http://thunder.msfc.nasa.gov/data/query/distributions.html)

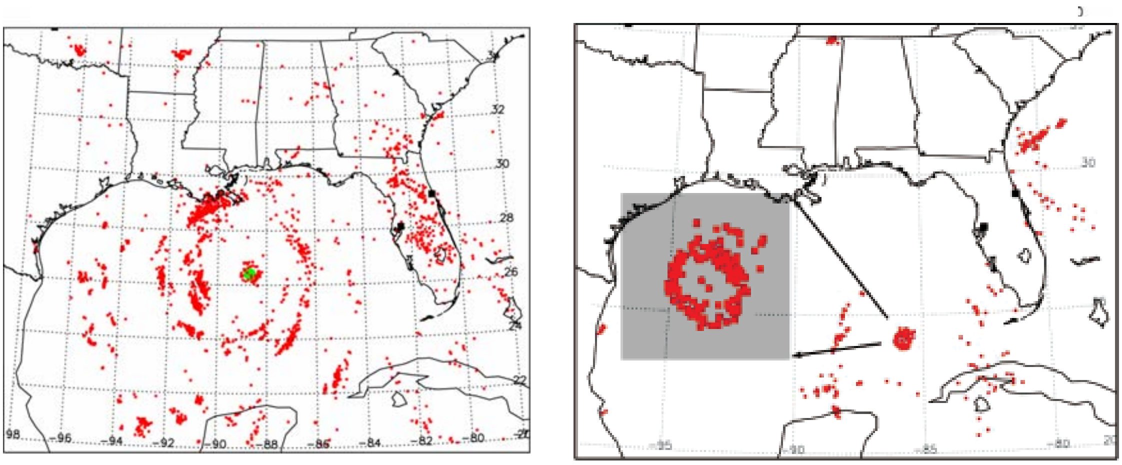

Figure 2. Eye-wall lightning density in hurricane Katrina when it progressed from Cat 4 to 5 (17:30-18:30 UTC, 28 Aug 2005) (left) and in hurricane Rita (right panel) during its intensification from Cat 3 to 5 (14–15 UTC, 21 Sep 2005) (right) (after Shao et al., EOS, 86, 42, 18 Oct. 2005)

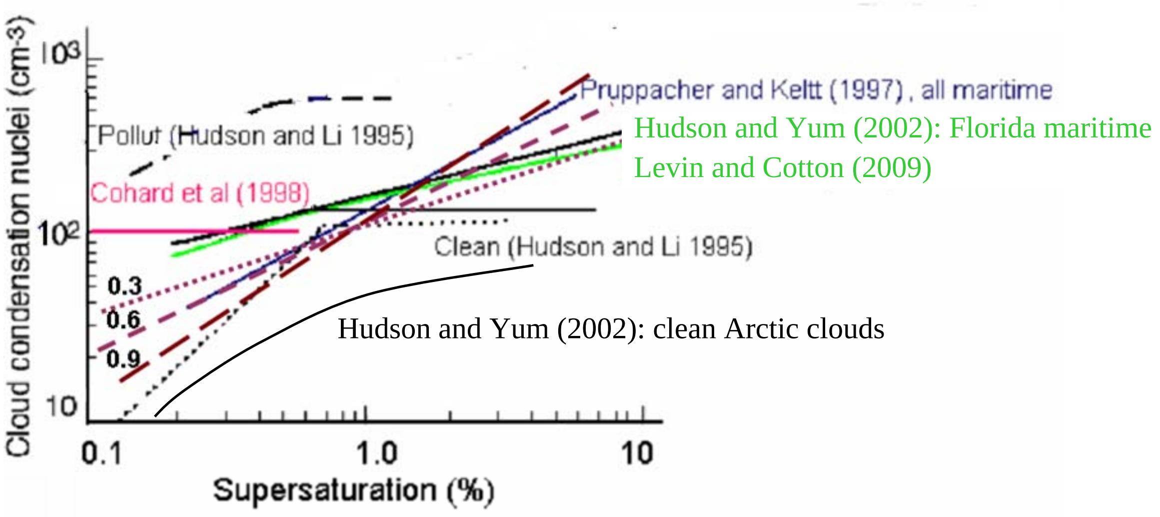

Figure 3. Dependences of concentration of activated CCN on supersaturation over the sea reported by different authors. Lines denoted 0.3, 0.6 and 0.9 correspond to dependencies (1) with corresponding values of parameter k. One can see that the dependence presented by Pruppacher and Klett (1997) for "all maritime" cases is close to the case k=0.9.

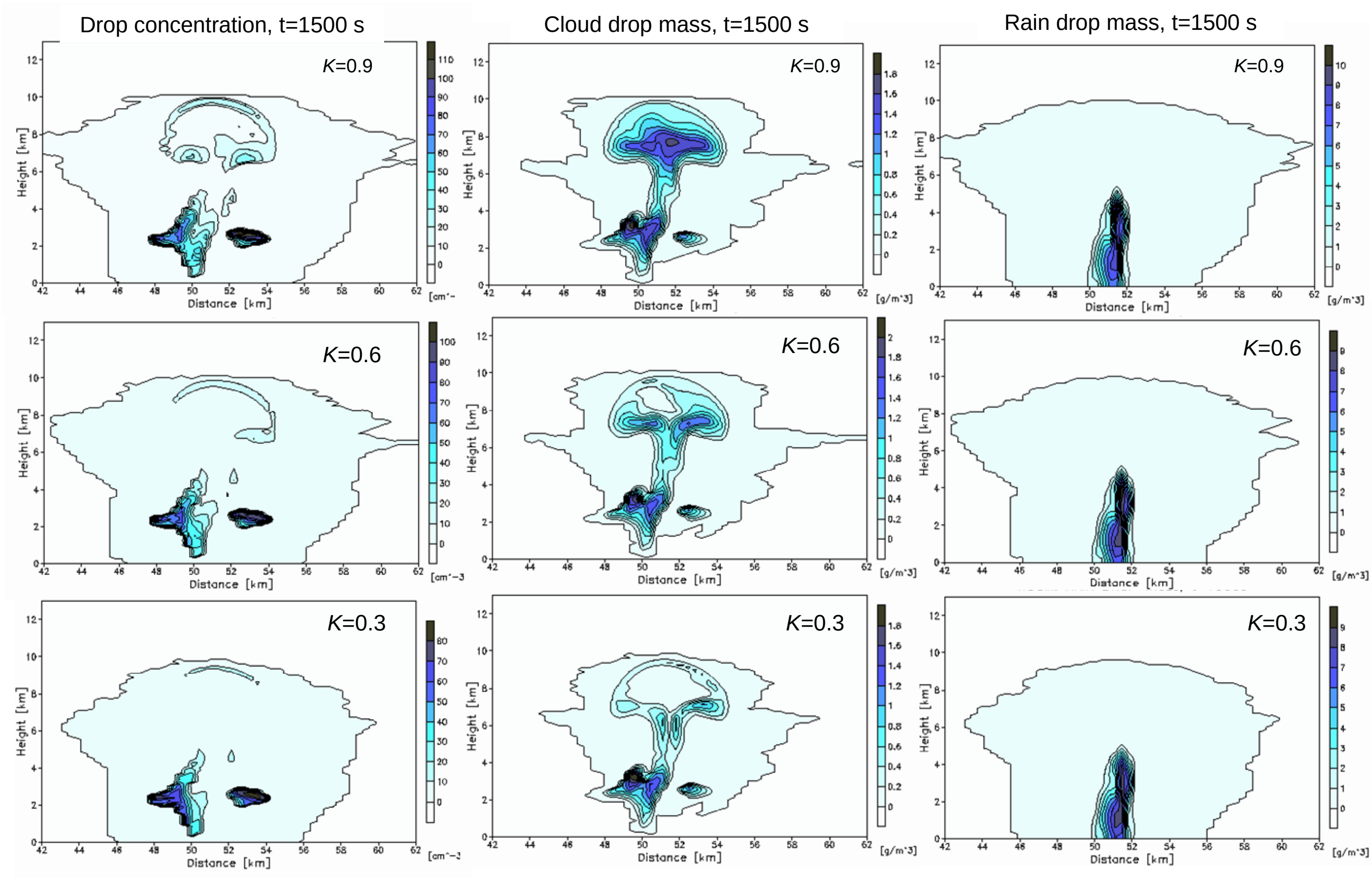

***Figure 4**. Fields of the droplet concentration N_d (left), the cloud water content (CWC) (middle), and rain water content (RWC)(right) for N_0=100 cm^{-3} at different slope parameters: k=0.9 (upper panels), k=0.6 (middle panels) and k=0.3(low panels).*

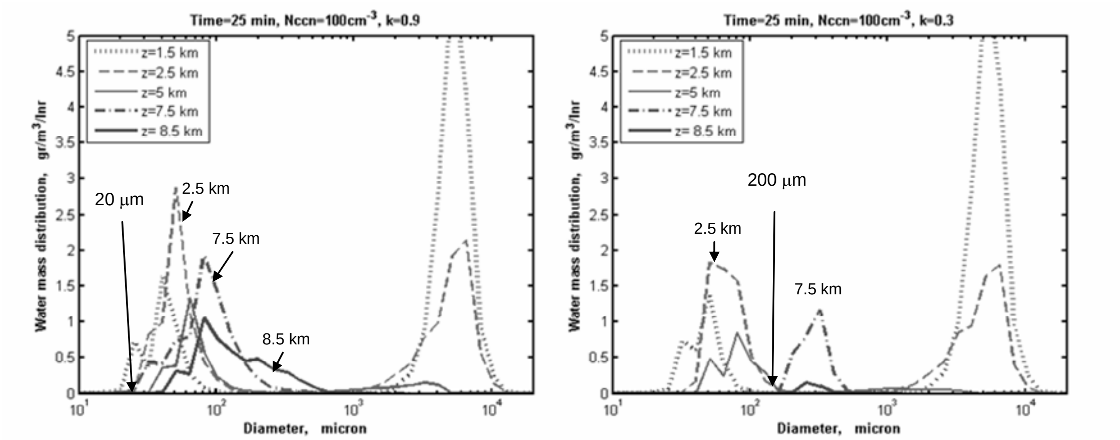

Figure 5. Droplet size distribution (DSD) at several height levels along cloud axes in the cases k=09 (left) and k=0.3 (right) at t=1500s. Numbers 20 μm and 200 μm indicate the minimum droplet size in the DSD at z= 7.5 km in the runs.

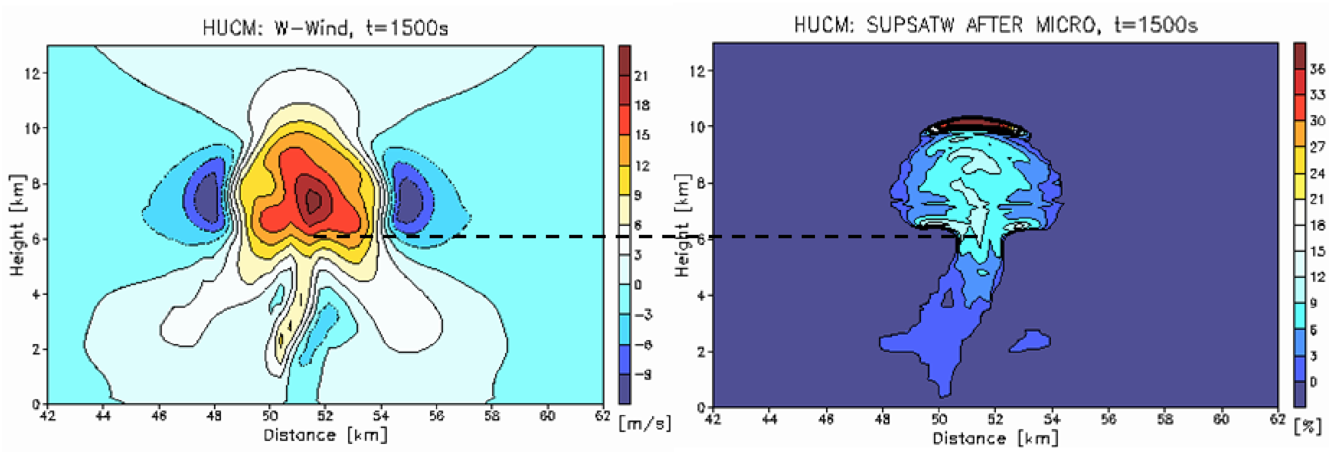

Figure 6. The fields of vertical velocity (left) and supersaturation (right) in simulation with k=0.9. The dashed line denotes the level of high acceleration of vertical velocity. One can see that increase in vertical velocity leads to excess in supersaturation over 12%.

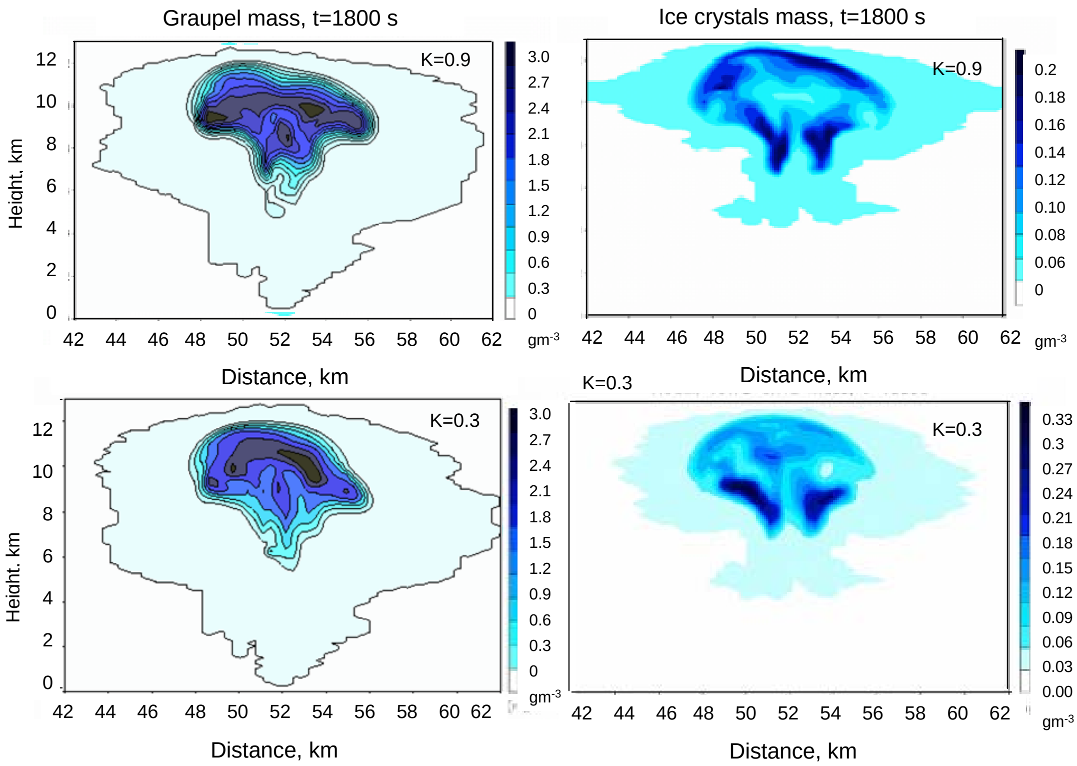

Figure 7. Fields of graupel (left) and ice contents (right) in the simulation with k=0.9 (upper row) and k=0.3 (low) at t=1800 s.